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Use of nanofiltration and reverse osmosis in reclaiming micro-filtered biologically treated sewage effluent for irrigation

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HIGHLIGHTS

- Nanofiltration (NF) pre-treatment reduced reverse osmosis (RO) membrane fouling.
- Permeate blends of RO after NF treatment and NF only are suitable for irrigation.
- NF or RO, alone removed most pharmaceuticals and personnel care products (PPCPs).
- PPCPs removals by NF membranes were lower than those by RO membranes.

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ABSTRACT

Micro-filtered, biologically treated sewage effluent (BTSE) generally has high sodium adsorption ratio (SAR) and sodium (Na) and chloride (Cl) concentrations. Therefore it cannot be directly used for irrigating sensitive crops. A study was conducted on a micro-filtered BTSE from a Sydney water treatment plant to determine whether the BTSE can be treated using nanofiltration (NF) and reverse osmosis (RO) to bring these risk parameters within safety limits. The study showed that using NF and RO alone could not produce the required ratio of SAR. Furthermore, NF alone did not remove the necessary levels of Na and Cl ions while RO did. However, blending equal proportions of NF permeate and RO permeate obtained from a two stages hybrid treatment system consisting of NF followed by RO resulted in a product quality suitable for irrigation in terms of the above mentioned risk factors. Utilizing NF prior to RO reduced the RO membrane fouling as well. Both NF and RO removed most of the pharmaceutical and personal care products from the feed water and this may subsequently protect soil and ground water from potential hazards.

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1. Introduction

Reclaimed wastewater for irrigation serves as an economical water resource in many countries [1]. It also has several benefits in improving soil health and reducing the need to use fertilizers. However, excessive salts, pathogens, trace organics, sodium (Na) and chloride (Cl) can cause dangerous environmental risks. The water quality criteria for irrigation are mainly characterized in terms of salinity and Na hazards, pH, and concentrations of some specific ions such as Cl^- , borate (BO_3^{3-}), and nitrate (NO_3^{-}).

Salinity is a hazard that results from high salt content in the water which directly affects plant growth, crop performance and soil properties [2] and it can be expressed by electrical conductivity (EC). High EC may cause physiological drought in plants. Sodium hazard is measured by sodium adsorption ratio (SAR) which provides the relative

* Corresponding author. *E-mail address:* s.vigneswaran@uts.edu.au (S. Vigneswaran). concentration of Na to calcium (Ca) and magnesium (Mg) ions. An excessive level of Na in relation to Ca and Mg affects the permeability characteristics of soil profile by changing the soil structure [3]. In addition to these, some specific ions such as Cl^- , BO_3^{3-} and NO_3^- at excessive levels can severely damage plant growth.

According to Ayers and Westcot [4] an excess concentration of Cl⁻ in soil solution causes this element to accumulate in plant leaves and cause leaf burn/dead leaves. This eventually results in necrosis (dead tissue). While boron (B) is an essential element for plant growth the high concentration of this element causes older leaves to turn yellow and this ultimately causes chlorosis. Nitrogen (N) is also an important element but its over-supply may over-stimulate plant growth, leading to delayed maturity of produce and ultimately its poor quality. As such, nutrient balanced irrigation water is essential in order to have a positive impact on plant growth. According to the water quality standards reported by ANZECC [3], the allowable safety limits of SAR, Cl, Na and B are 2–8, <175 mg/L, <115 mg/L, and <0.5 mg/L for very sensitive crops. The desirable range of pH for irrigation water is 6.5 to 7.6. The pH beyond this







range (due to bicarbonates and carbonates) causes Ca^{2+} and Mg^{2+} ions to form insoluble precipitates and consequently Na^+ ions become dominant.

However, these standards may vary depending on the sensitivity of crops, SAR and EC of the water, and soil type. Besides these inorganic constituents, pharmaceuticals and personal care products (PPCPs) in irrigation water are increasingly accumulating in crop tissues and this has important implications for people's health upon consumption. PPCPs are contaminants that have the properties of toxic biological hazards even at low concentrations. Carter et al. [5] reported the accumulation of some pharmaceuticals in the tissues of radish (*Raphanus sativus*) and ryegrass (Lolium perenne). Another study reported the presence of pharmaceutical residues in plants tissues (especially for alfalfa and apple) which were irrigated by reclaimed water containing pharmaceuticals [6]. The long-term use of irrigation water containing PPCPs may eventually lead to potential groundwater contamination. The occurrence of PPCPs in groundwater has been documented in some studies over the last decade [7–9]. However, the critical toxic values for most of the PPCPs have not been reported in the literature.

Membrane technologies play a key role in reclaiming micro-filtered biologically treated sewage effluent (BTSE) and have received much attention during the past few decades owing to the need to overcome water shortage problems [10]. Studies have mainly investigated combining membrane filtration (MF) and ultrafiltration (UF) with RO membranes to remove suspended particles as well as to reduce salinity levels [11,12]. Bunani et al. [2] used RO technology to treat biologically treated sewage effluent (BTSE) for irrigation and suggested blending 20-30% of BTSE and 80-70% of RO permeate to make product water suitable for irrigation. However, it is not economical to blend high volumes of RO. Mrayed et al. [13] reported a combination of NF and RO treatment processes to treat BTSE and recommended a blending of NF concentrate and RO permeate for irrigation. The reason for this particular blending was to enrich the product water with divalent nutrients as well as to reduce monovalent nutrients in the product water because NF has the ability to reject divalent ions. Conversely, RO can reject both monovalent and divalent ions [14]. They suggested blending NF concentrate and RO permeate at the ratio of 32:68 which resulted in a SAR of 8.2 but this resulted in a high concentration of Na ions (588 mg/L) which is not suitable for Na sensitive crops.

None of the above studies have investigated the removal of PPCPs along with inorganics from BTSE water for irrigation use. The objective of this study was to evaluate combining NF and RO (a two stages hybrid system) to raise the quality of micro-filtered BTSE water in terms of SAR value and Na and Cl concentrations so that it was suitable for irrigation. The possibility of using NF followed by passing part of the NF permeate through RO and combining the NF and RO permeates at suitable ratio to achieve good irrigation water quality was tested. The product water's

Table 1

Physico-chemical characteristics of feed water.

quality was also evaluated for PPCPs to prevent them from poisoning groundwater and soil over the long-term. Furthermore, the layout/configurations of NF and RO membranes were investigated in terms of reducing potential RO membrane fouling.

2. Materials and methods

2.1. Materials

2.1.1. Feed water

The micro-filtered BTSE collected from a water reclamation plant located in Sydney, Australia was used as feed water. Its characteristics and water quality criteria for irrigation use are presented in Table 1. The use of this feed water itself is unsuitable for sensitive crops as the SAR value was 39, and levels of Na⁺ and Cl⁻ were 81–120 mg/L and 150–300 mg/L, respectively. Therefore the feed water needs to be further treated.

2.1.2. Membranes

Three types of NF membranes and an RO membrane were used in this study to compare their effectiveness in removing contaminants of concern. The characteristics of the membranes are presented in Table 2. These three membranes were selected because of their differences in zeta potential or molecular weight cut off (MWCO) value or both, which would help in identifying the mechanisms of DOC, salts and PPCPs removals.

2.2. Methodology

A known quantity (20 L) of micro-filtered BTSE was filtered through NF or RO membrane (Fig. 1). The NF and RO filtration units (Fig. 1) were equipped with a rectangular cross-flow cell having a membrane area of 68 cm². The membrane charge has been shown to become less negative (reduced zeta potential) when the temperature of the feed water increased [18]. Therefore, a cooling coil was submerged in the feed water tank to maintain the feed water temperature at a constant 20 \pm 2 °C. A pressure of 4 bar was used for all NF membranes. The clear water fluxes $(L/m^2 \cdot h)$ were 55, 12, and 62 for NP 010, NP 030, and NTR 729HF, respectively. Thus the corresponding clear water permeabilities $(L/m^2 \cdot bar \cdot h)$ were 13.75, 3 and 15.5. The pressure used for RO was 40 bar. The clear water flux was 23.5 $L/m^2 \cdot h$ and the clear water permeability was 0.59 $L/m^2 \cdot bar \cdot h$. The concentrate (retentate) produced from NF or RO was recirculated back into the feed water. The performance of each membrane was tested using the same operating conditions of the membrane unit. Of the three types of NF membranes the best one was selected for combining with a RO posttreatment.

Parameter	Unit	Micro-filtered BTSE	Australian and New Zealand Guidelines for Fresh and Marine Water Quality [3]
Dissolved organic carbon (DOC)	mg/L	3.6-7.7	
рН	-	6.8-7.6	6.5-8.0
conductivity	dS/cm	0.52-1.12	<0.65; 0.65–2.9; 2.9–5.4; >8.1 for very sensitive; sensitive to moderately tolerant; tolerant to very tolerant; too saline.
SAR		39	2–8; 8–18; 18–146; 46–102 for very sensitive; sensitive; moderately tolerant and tolerant crops.
F	mg/L	0.7-1.1	1.0 and 2.0: long term trigger value and short term trigger value
Cl ⁻	mg/L	150-300	<175; 175-350; 350-700; >700 for very sensitive; sensitive; moderately tolerant and tolerant crops
NO_3^-	mg N/L	1.0-1.3	5; 25–125 for long term trigger value and short term trigger value
PO ₄ ³⁻	mg P/L	0.74-0.99	0.05; 0.8–10 for long term trigger value and short term trigger value
SO ₄ ²⁻	mg S/L	49-51	
Na ⁺	mg/L	81-120	<115; 115–230; 230–460; >460 for sensitive; moderately sensitive; moderately tolerant and tolerant crops.
K ⁺	mg/L	15-21	
Ca ²⁺	mg/L	21-40	
Mg ²⁺	mg/L	10-15	
BO ₃ ³⁻	mg B/L	0.04-0.06	<0.5; 0.5–2.0; 2.0–6.0; 6.0–15.0 for sensitive; moderately sensitive; moderately tolerant and tolerant crops.

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